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EXHIBIT A

LYNNTECH, Inc.

Patent Disclosure No. _____

Title of invention:

Novel Separator Concepts for Electrochemical Cells

Inventors:

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Clarke

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Novel Separator Concepts for Electrochemical Cells

Prior Art

Figure 1 is a cross section view of an idealized proton exchange membrane (PEM) fuel cell. This single cell illustrates all of the key features in the cell and includes the key fuel cell reactions for a hydrogen-air fuel cell. For practical applications a series of such cells are assembled in series in what is known as a stack. In that situation, each anode and cathode is not a discrete part, as shown in Figure 1. Instead each part has an anode flow field or flow field attachment point on one side and a cathode flow field or flow field attachment point on the other, making the plate bipolar. These bipolar plates represent the area of this invention.

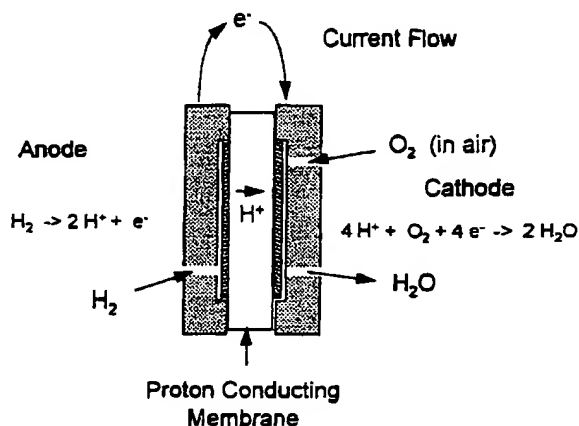


Figure 1. A schematic cross section of a PEM fuel cell illustrating the key features. The thickness of the membrane has been greatly exaggerated for clarity.

The existing technology for PEM fuel cells utilizes a variety of materials of construction for the bipolar separator plates. These materials include non-metallic conductors, such as graphitic carbon,¹ as well as a variety of metals, including titanium and stainless steel.² In all cases, the bipolar plate is fabricated by machining the flow fields into a solid sheet of material, as illustrated in Figure 2.

While this is a relatively straight forward approach, it does have several disadvantages. The first of these is density. Consisting of a solid piece of graphite or metal limits the density of the final part to close to the density of the original

stock, since the machining rarely removes more than about $\frac{1}{4}$ of the material. The second disadvantage is price. Machining each piece from a solid starting blank means that each piece requires the use of relatively expensive machining processes, instead of less expensive molding, casting or stamping processes.

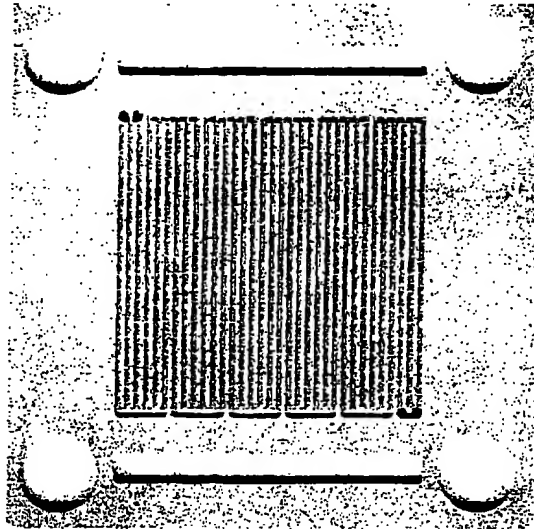


Figure 2. A view of a typical metal separator plate with a serpentine flow field design. This design features three parallel channels along each leg with a common manifold connecting each set of channels to the next.

Abstract of the Invention

This invention is an improved separator plate design, based on the use of lighter weight materials and simpler, less expensive, more easily automated manufacturing processes.

Reducing the weight is achieved by minimizing the amount of dense material (solid metal or graphite) and replacing it with lighter materials. Solid material is only used where it is absolutely necessary, such as in the outer frame of the bipolar plate where a solid structure and a gas seal are required. Other parts of the plate, such as the flow field, are constructed from less dense materials, such as expanded metal mesh (Figure 3), or foamed metal (Figure 4).

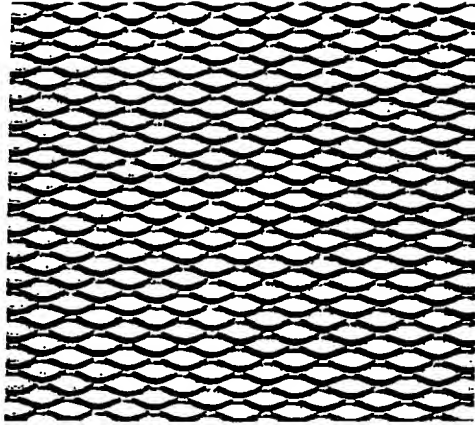


Figure 3. Magnified view of a sheet of expanded metal. Expanded metal is produced from a metal sheet by making a series of small cuts and stretching the sheet to expand it. Most metals available in sheet form can be used to produce expanded metal.

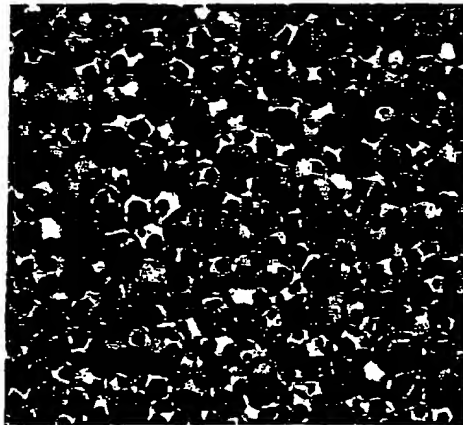


Figure 4. Magnified view of a sheet of foamed metal (copper). Foamed metal is produced by one of two proprietary methods, using either a polymer foam template process or by inert gas blowing of a melt. Copper and nickel foams are produced by the former route and aluminum foams by the latter.

As is readily apparent from the two figures above, these materials are not gas tight, and if used alone would not function successfully as a gas barrier. The gas barrier can be produced in one of two ways. The first of these is to use a metal sheet as the gas barrier. This approach is illustrated in Figure 5. In this embodiment foamed metal or expanded is bonded to either side of the barrier, so

that the low density metal serves as the gas distributing flow field and the metal sheet serves as the barrier. This barrier can be made far thinner than any gas barrier produced by machining away material to leave a flow field and barrier structure made from a single piece of material.

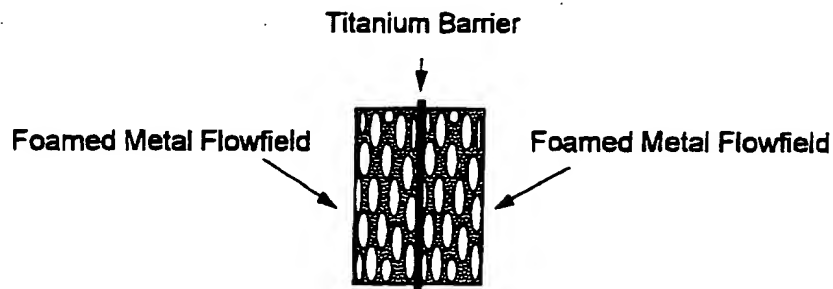


Figure 5. Cross-section of an all metal lightweight bipolar plate. This design features a thin ($\leq 0.006"$) barrier made from solid titanium sheet with foamed metal flow fields welded to both sides of it.

Another alternative is to use produce a polymer barrier inside a solid piece of low density metal. This method has some additional advantages, by using materials with special properties, such as water permeability, for the gas barrier, it is possible to impart the separator plates with additional advantages.

Examples

The following examples show the function of this invention and some of its preferred embodiments.

1. **This example shows the performance of a two cell stack produced using an all metal bipolar plate.**

A bipolar plate was produced by first mounting a sheet of 0.006" (0.15 mm) in a lightweight (aluminum) frame produced from the design shown in Figure 6 using aluminum 0.125" thick. (An alternative design for this component, with a different flow orientation, is illustrated in Figure 7.) This metal sheet serves as the gas barrier.

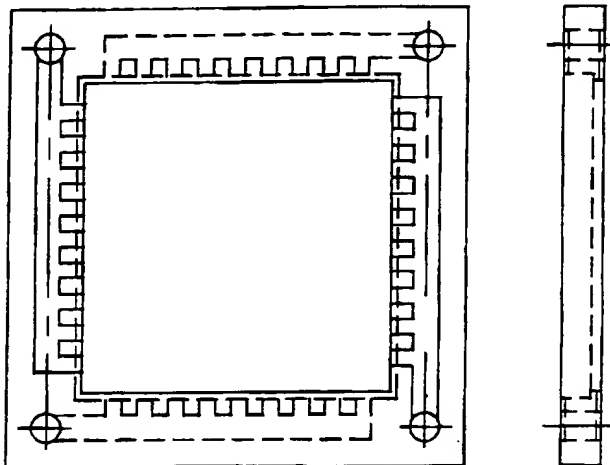


Figure 6. CAD drawings for a $\frac{3}{16}$ " thick frame for a bipolar plate. The same design can be used in thicknesses from $\frac{1}{8}$ " to $\frac{1}{4}$ ".

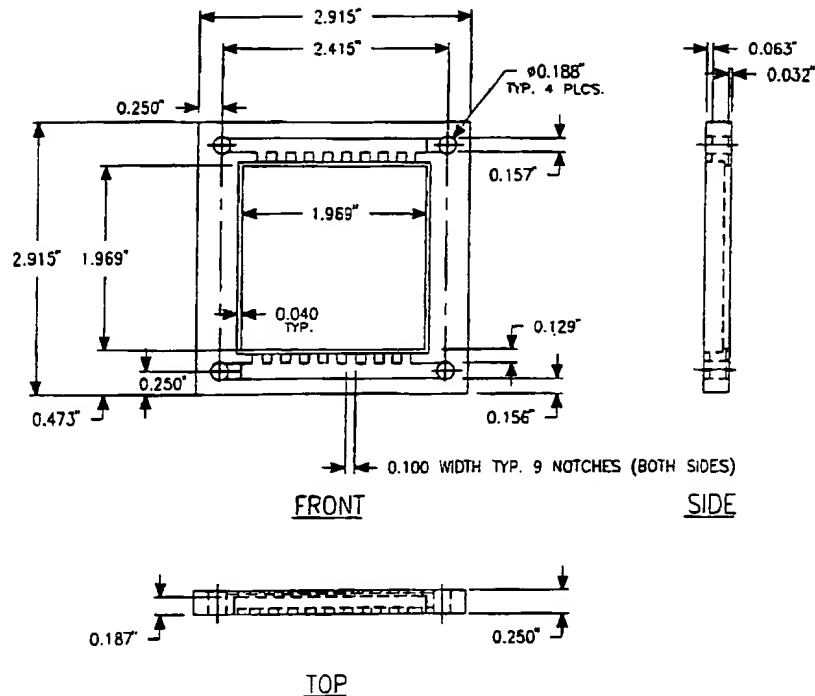


Figure 7. CAD drawings for an alternate design to the one shown in Figure 6 for the frame of a bipolar plate. In this design the gas flows are parallel or opposing rather than the cross flow arrangement. Like the one in Figure 6, this frame can be produced with a thickness anywhere between $\frac{1}{8}$ " and $\frac{1}{4}$ ".

Flow fields for gas distribution are produced by welding pieces of expanded titanium (as seen in Figure 3) to either side, as illustrated in Figure 5. Since the gas distribution is cross flow, the two pieces are oriented at 90° to each other with the long axis of the diamond pattern in-line with the flow on each side. After attachment of the flow fields, the titanium was gold plated to prevent the formation of an insulating layer of titanium dioxide on the surface of the flow field.

The bipolar plate was used to assemble a stack which was operated with the results shown by the polarization curves in Figure 8. After this data was obtained the stack was disassembled and examined. There was no evidence of corrosion on any of the parts, as shown in Figure 9.

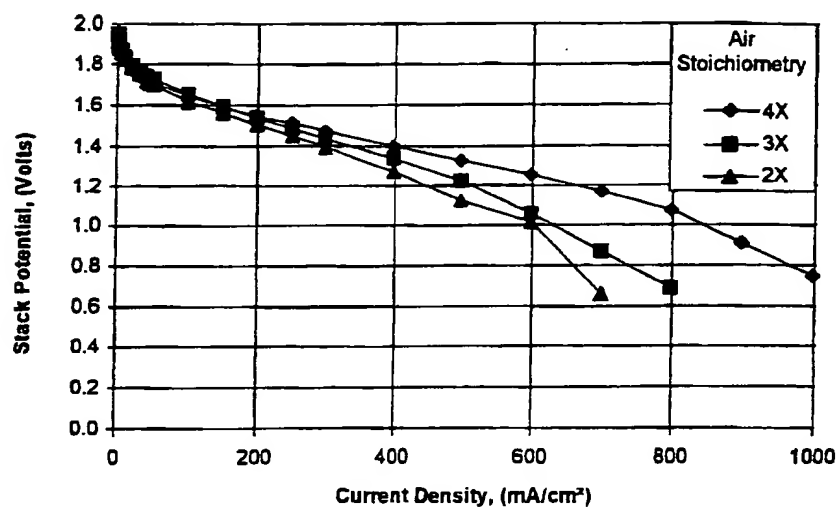


Figure 8. Polarization curves for a two-cell stack using a bipolar plate with expanded titanium flow fields operating at 75 °C and 30 or 36 psig pressure at three air stoichiometries.

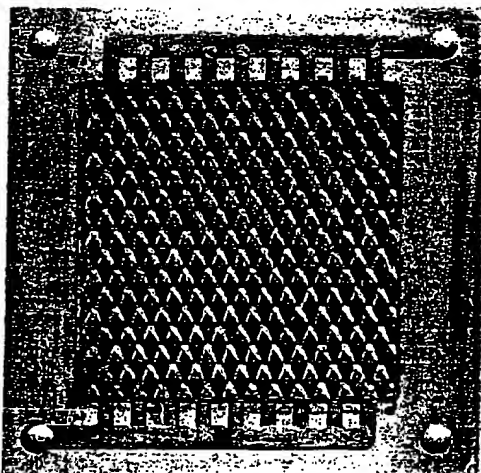


Figure 9. Used bipolar plate constructed by spot welding expanded titanium flow fields to both sides of a titanium barrier layer. The titanium was gold plated after welding to prevent oxidation of the metal in the fuel cell environment.

2. This example shows an alternative approach to an all metal bipolar plate.

A second bipolar plate was produced, again using a frame produced from 0.125" aluminum like that shown in Figure 6 and again bonding a sheet of 0.006" titanium into the frame as a gas barrier. For this bipolar plate foamed copper, like that shown in Figure 4, was welded to the titanium barrier and gold plated.

A two cell stack was produced using the bipolar plate, and the stack operated under a variety of conditions. The stack's performance is documented Figure 10. This performance is clearly an improvement from the results described in Claim 1 and illustrated in Figure 8.

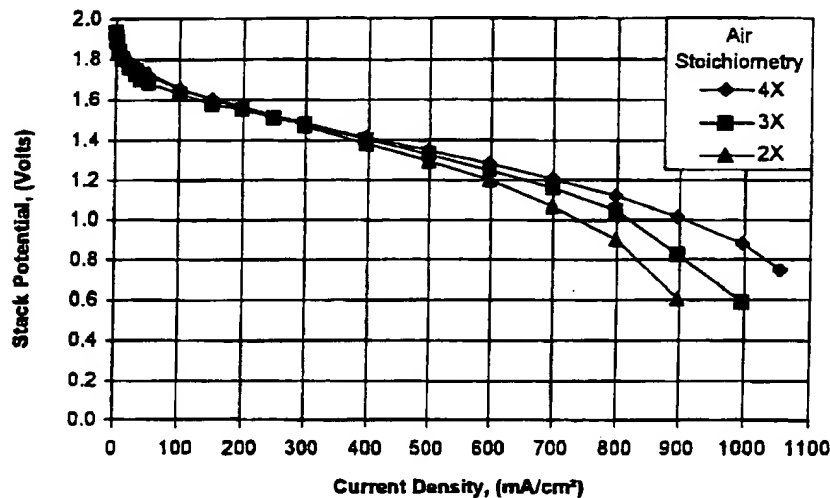


Figure 10. Polarization curves for a two-cell stack using a bipolar plate with gold plated foamed copper flow fields operating at 75 °C and 36 psig pressure at three air stoichiometries.

At the conclusion of testing, the stack was dismantled and the components inspected. As shown in Figure 11, there was no visible attack or degradation of the bipolar plate.

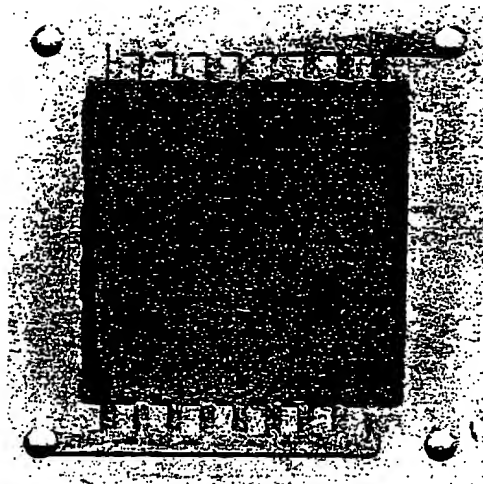


Figure 11. Used bipolar plate constructed by spot welding foamed copper flow fields to both sides of a titanium barrier layer. The foamed copper was gold plated after welding to prevent oxidation of the metal in the fuel cell environment. The gold plating on the aluminum frame is spill-over from the plating of the copper foam. The darkening at the edges is material from the back of the electrode.

3. This example shows a bipolar plate with a polymeric barrier.

As described above, it is also possible to produce a bipolar plate with a polymer barrier. In this case, a sheet of foamed metal, as shown in Figure 4, is partially impregnated with epoxy and the epoxy cured. For this example, the resin used was Epon™ 862 (Shell Chemical), and the hardener was Epicure™ (also Shell). A cross section of such a plate appears in Figure 12.

The foam with barrier is assembled into a bipolar plate by first sealing the filled plate into a $\frac{3}{16}$ " aluminum frame like that shown in Figure 6. The filled plate is sealed into the frame against the inner support ridge machined into the inner pocket of the frame. The plate is completed by attaching a thin, unfilled piece of foamed metal to the opposite side. A cross section of the finished assembly is illustrated in Figure 13. The calculated densities of the barrier regions of plates made with epoxies from two major families and several types of metal foam are shown in Table I.

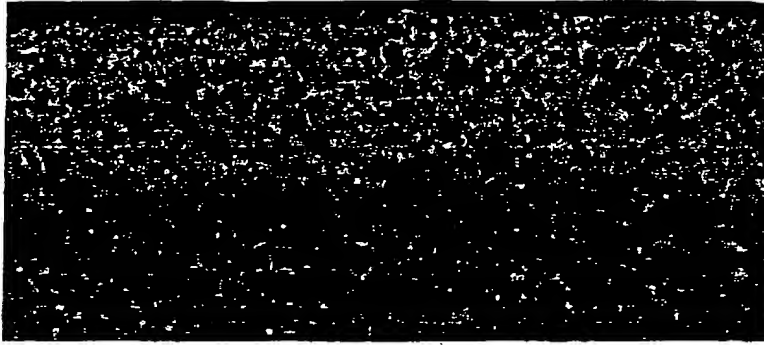


Figure 12. Magnified cross-section of a sheet of copper foam with an epoxy gas barrier. The filled portion of the film is at the bottom of the picture.



Figure 13. Enlarged cross section of the improved plate frame showing how the polymer filled metal foam fits into the plate. The patterned section is the polymer filled metal foam, with the lower piece being the piece with the gas barrier inside of it. The gas barrier is located near the center of the plate.

This plate was used to assemble a two cell fuel cell stack. The stack was operated using hydrogen as fuel and air as the oxidizer, with the results shown in Figure 14 for ambient pressure use and Figure 15 for pressurized operation. These results do not show performance equal to that shown in Figure 10.

Table I. Densities of Epoxy Filled Metal Foam Composites

		Metal Component			
Bisphenol A types	1.16	1.94	1.55	1.31	1.24
Bisphenol F type	1.23	2.00	1.61	1.38	1.30

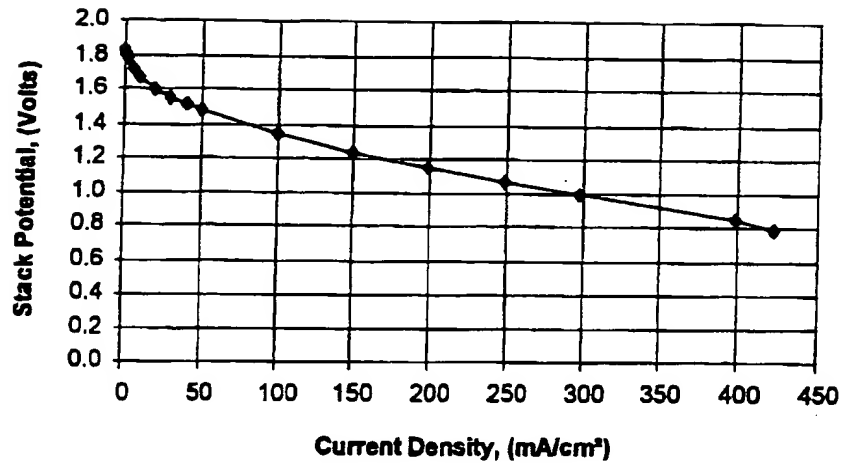


Figure 14. Polarization curve for a two-cell fuel cell stack equipped with an Epon 862 filled copper bipolar plate operating on hydrogen air at ambient pressure and 55 °C.

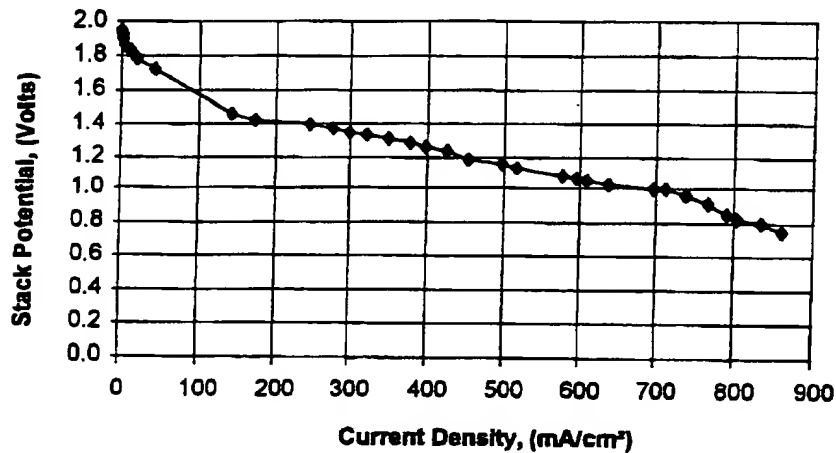


Figure 15. Polarization curve for a two-cell fuel cell stack equipped with an Epon 862 filled copper bipolar plate operating on hydrogen air at 30 psig pressure and 65 °C.

4. This example shows a polymeric gas barrier produced using a thermoplastic.

The same type of structure, metal foam with a polymer gas barrier can be fabricated using thermoplastic materials. Figure 16 shows the fabrication procedure. A variety of polymers have been tested. In each case the polymer was heated to a temperature above the published softening point. The temperatures used for three polymers tested are tabulated in Table II.

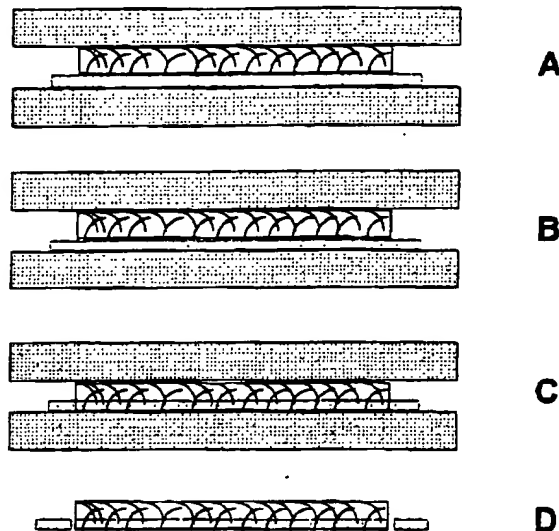


Figure 16. Impregnation procedure used for thermoplastic barrier materials. The dark gray rectangles are the platens of the press, with the metal foam shown with a herring bone crosshatch and the polymer in light gray. The steps illustrated are: A Place foam in press on top of a sheet of thermoplastic, B Heat press until the plastic melts, C Press foam into molten polymer, and, D Cool and trim off excess polymer.

Table II. Press and Transition Temperatures for the Polymers Under Test

Polymer	$T_{(press)}$ (°C)	T_g (°C)
PES	343	232
PC	288	285
Nylon	268	246

The press temperatures listed in the table are those that were experimentally determined to give the best results. All of the data included here are for polymers which produced plates that withstood at least 20 psig gas pressure. The degree of penetration achieved for each of the polymers can be clearly seen in the figures below. One of important feature for producing a good gas barrier is the ability to form an even layer.

Figure 17 shows the results for polyethersulfone (PES). This polymer produces the cleanest demarcation of any of those examined. Figure 18 shows a polycarbonate (PC) barrier. It produces a good barrier layer but also shows signs of deeper penetration into the foam. The last of these figures, Figure 19, shows a Nylon 6 barrier. This is the roughest in appearance of the three shown here.

Forming the gas barrier with a thermoplastic polymer by hot pressing has the potential for easy automation and rapid production. With the thermoplastic there is no cure time, only the cooling time, and the cooling time may be as short as seconds.

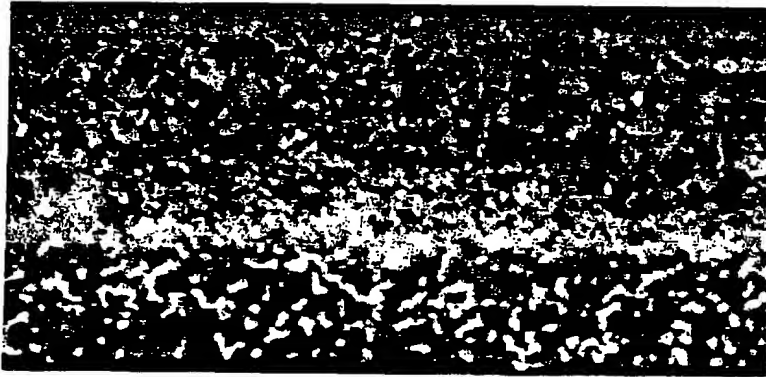


Figure 17. Magnified (20X) cross section of a foamed copper specimen filled with PES. Note the clear demarcation between the filled region and the open region.

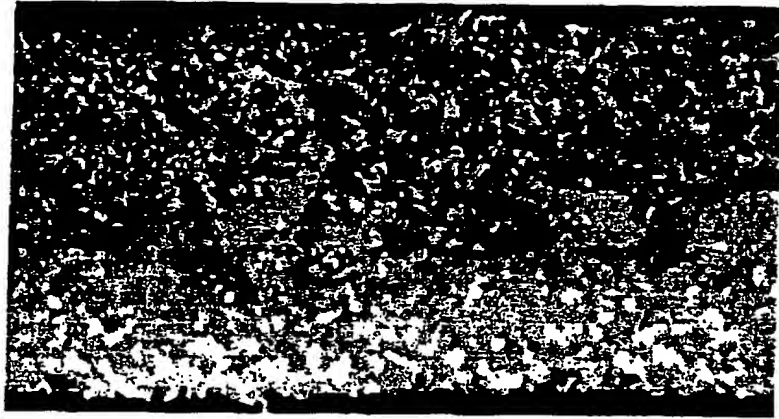


Figure 18. Magnified (20X) cross section of a foamed copper specimen filled with PC. Note that the demarcation between the filled region and the open region is less clearly defined than it was with PES.

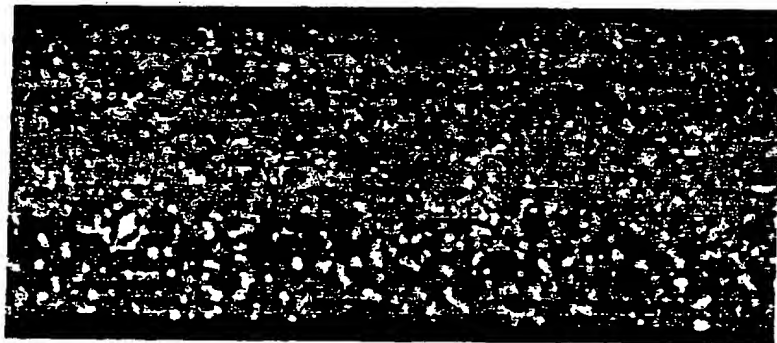


Figure 19. Magnified (20X) cross section of a foamed copper specimen filled with Nylon 6. Note that the demarcation between the filled region and the open region is poorly defined with fingers of polymer extending into the clear portion of the foam.

Table III. Densities of Polymer Filled Metal Foam Composites

Polymer	d. (g/ml)	Metal Component			
		Cu, 10%	Cu, 5%	Al, 10%	Al, 5%
Polyethersulfone	1.37	2.13	1.75	1.50	1.44
Polyethylene Terephthalate	1.36	2.12	1.74	1.49	1.43
Cellulose Acetate	1.30	2.06	1.68	1.44	1.37
Polycarbonate	1.20	1.97	1.59	1.35	1.28
Nylon™ 6	1.13	1.91	1.52	1.29	1.21
HDPE	0.95	1.75	1.35	1.13	1.04

5. This example shows how to make a water permeable bipolar plate. Using a polymer gas barrier in a metal foam matrix makes a relatively lightweight bipolar plate. These plates can be given additional properties if the polymer has special properties.

The normal reactions occurring in a fuel cell are shown in Figure 1. As shown, water is formed in the cathode by the reduction of oxygen. Water is also transported to the cathode electroosmotically from the anode side of the membrane as each proton drags an average of two water molecules with it. This electroosmotic drag will eventually dry out the membrane, leading to higher internal resistance, and lower efficiency, unless it is replaced. Simple back diffusion from the cathode to the anode is generally not enough to replace the electroosmotically moved water, even though water is being formed continuously at the cathode.

The usual solution to this problem is to supply additional water as water vapor in the fuel stream, but this requires the presence of an additional part, a humidifier, somewhere in the system.

If the bipolar plate is water permeable, there is no need for any additional humidification. The high concentration of water present at the cathode, relative to that present at the anode, furnishes the driving force for diffusion, and water diffuses through the water permeable bipolar plate from the wet side (cathode) to the dry side (anode) where it evaporates into the fuel stream and is available to enter the membrane.

Using a water permeable polymer, such as Nafion™ or cellulose acetate, to construct the gas barrier in a bipolar plate like those described in Example 4 makes the bipolar plate water permeable and eliminates the need for any additional humidification.

Figure 20 shows the flows in a portion of a fuel cell stack produced based on this approach.

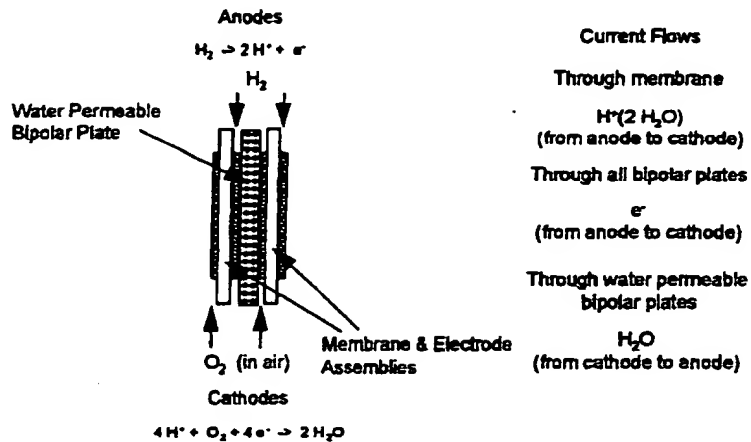


Figure 20. Flows in a fuel cell stack, demonstrating the advantages of a water permeable bipolar plate.

6. This example shows bipolar plate with a polymer barrier and provision for cooling.

Examples 4 and 5 show how to produce lightweight bipolar plates with polymer gas barriers. While these may represent an improvement on previous methods of producing bipolar plates, they do not solve all of the problems in fuel cell design.

A major challenge in operating a fuel cell stack at high current density is removing the waste heat generated by the stack. It must be remembered that even when a stack is operating at 50 % efficiency for the conversion of the thermal potential in the fuel to electricity (an efficiency above that achieved by most other conversion systems), the stack is generating as much waste heat as it does electricity. Therefore, the stack needs some method of removing the waste heat from the stack.

The most common method for doing this is the inclusion of dummy cooling cells in the stack. These cells, which can occupy as much as half of the total volume of the stack, serve to permit carry a coolant, or heat exchange fluid (typically water or an aqueous solution of a non-ionizing compound) through the cell to absorb the excess heat and carry out of the stack.

An alternative approach to solving this problem is to provide provision for coolant flow within the bipolar plates themselves. With most fabrication techniques for bipolar plates, this is difficult. With the method taught here for the fabrication of light weight bipolar plates with polymeric gas barriers, it is not difficult.

Figure 21 shows a cross section of a bipolar plate with provision for coolant flow. Where the use of dummy cells adds a volume equivalent to an entire cell to the stack for each cooling plate, this method only adds about 1 mm to the length of the stack. This is a definite improvement over current technology.

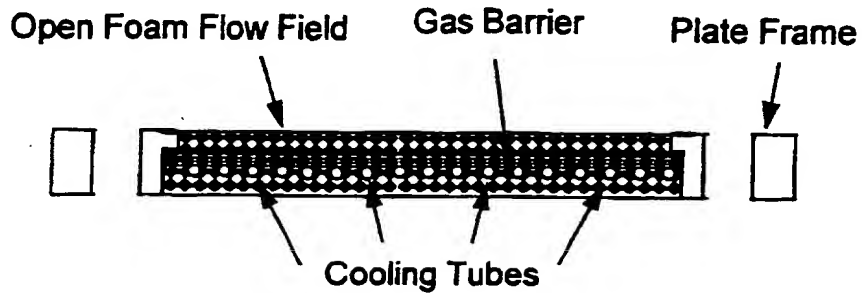


Figure 21. Cross section of a water permeable bipolar plate with hollow tubes inserted for cooling. In order to insure good water transport, the tubes must be kept to the minimum number needed for cooling.

7. This example shows a more advanced all metal bipolar plate. The bipolar plate designs taught in Examples 1 and 2 can be further reduced in weight while at the same time reducing the number of individual parts. The designs taught in those examples depended a metal frame around the lightweight flow field. Plates made from this design require a gasket to insure a gas-tight seal. (In some instances, the extension of the membrane outside of the active area is an adequate gasket.)

Figure 22 shows a partial cross section of a lighter version of this design. In this variation the metal gas barrier has been extended to the edges of the stack, and the metal frame around the perimeter replaced by a pair of relatively thick polymeric gaskets, one on each side of the gas barrier. This design reduces the weight by substituting a lower density polymer for the metal frame, and eliminates the need for any thin gaskets as are normally used between the metal frame and the membrane.

Specifically, two pieces of expanded titanium with an overall thickness of $\frac{1}{32}$ " (0.79 mm) are welded to opposing faces of a sheet of 0.006" (0.15 mm) titanium sheet. The entire assembly is prepared and gold plated to prevent corrosion and to improve electrical contact. This is assembled using 0.035" (0.89 mm) filled PTFE gaskets. The gaskets, as represented by a typical design in Figure 23, furnish sealing and gas channeling between the manifold ports and the expanded titanium flow field.

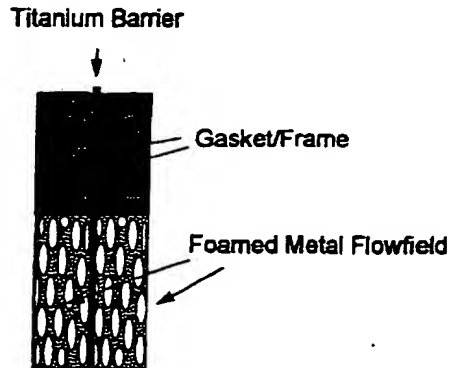


Figure 22. Partial cross-section of a bipolar plate based on a metal gas barrier with metal flow fields and polymer cell frames, which also serve as the gasketing.

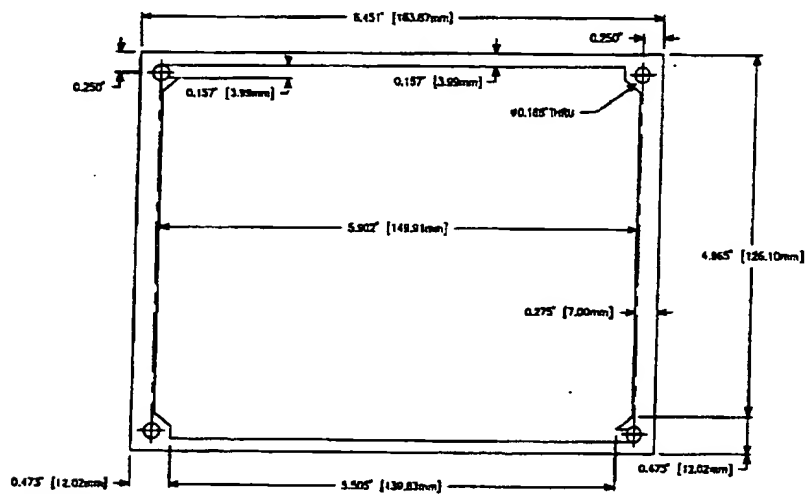


Figure 23. Face-on drawing illustrating the design for a polymeric frame/gasket for use in a stack built as described in Example 7 and illustrated in the partial cross section shown in Figure 22.

8. This example shows a method for enhancing gas distribution in metal foams.

When foamed metal is used as the flow field material, the random pattern of holes in the metal is used to distribute the gas. It is possible to improve the distribution of the gas flow, and reduce the pressure required to move the gas through the stack at the same time. This can be accomplished by milling an interdigitated groove pattern in the face of the metal foam, as illustrated in Figure 24. The shallow grooves, typically less than half the thickness of the foam, furnish reduced flow resistance paths to channel the gas over the entire surface of the electrode. The gas flows through the foamed metal segments between the grooves to reach the electrodes. To insure even distribution of gas, it is necessary to insure that the grooves from opposing sides of the flow field do not intersect.

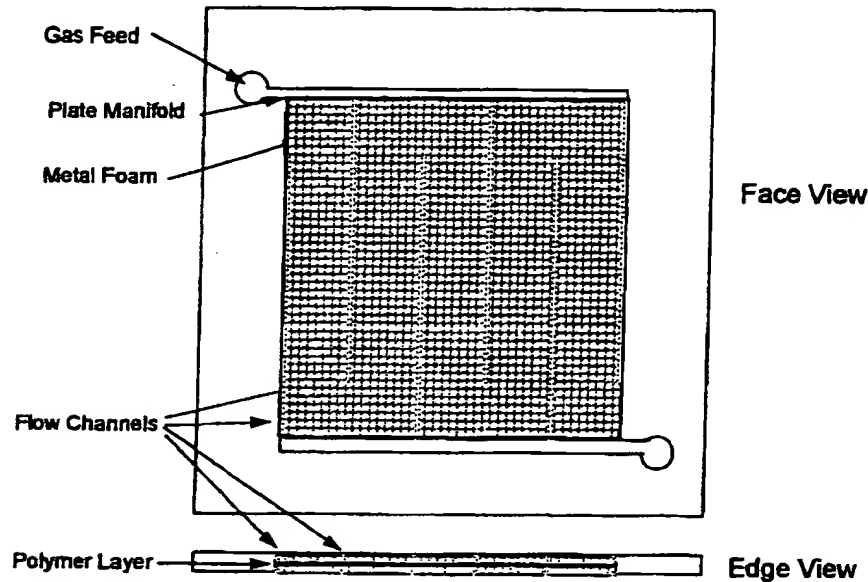


Figure 24. Face and edge views of a polymer modified metal foam bipolar plate illustrating the interdigitated flow field concept. This pattern of grooves milled into the surfaces of the foam are intended to improve the gas distribution at the surface of the electrodes and reduce the pressure drop. Note that the grooves do not intersect, thus forcing the flow through the foamed metal. (The feed and return holes for the second gas have been omitted from this drawing.)

9. This example shows a method for producing composite separator using polymers and a metal foam.

A gas barrier is constructed within a foamed metal sheet, as described in Examples 3 and 4. The difference in this case is that the gas barrier extends a significant distance beyond the edges of the central active area. In a separate step, a seal region is constructed at the perimeter of the plate by filling the foam with additional polymer. In the last step, the gas flow holes and grooves are added and the faces of the seal region smoothed. This produces a lighter bipolar plate that those produced with a metal frame.

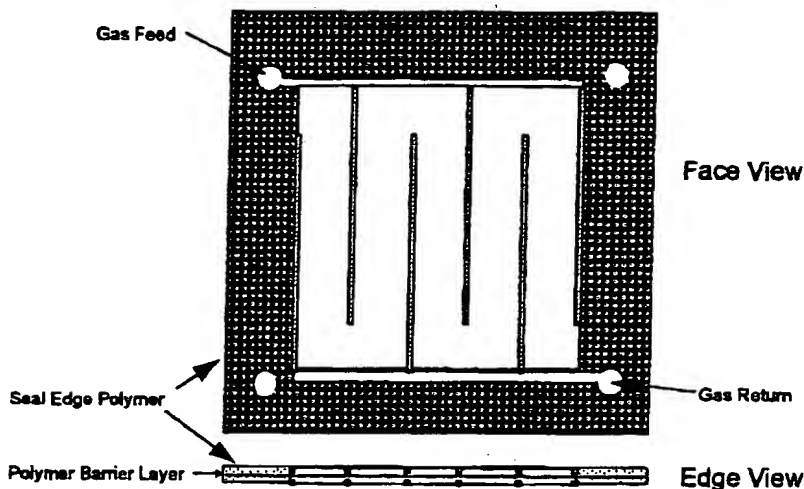


Figure 25. Two views of a bipolar separator plate produced by first constructing a gas barrier within a piece of foamed metal, then constructing a sealing barrier around the perimeter, and finally, adding the holes and groove structure.

Claims

We claim:

- 1.) A fluid separator plate useful in electrochemical cells having a metal barrier with a metal component formed to insure the even distribution of a fluid flow over the central area of the separator attached to on face and a second metal component formed to achieve the same type of distribution on the other face.
- 2.) The object of Claim 1 where the fluid distribution components are identical in structure.
- 3.) The object of Claim 1 where the fluid distribution components are differ in structure.
- 4.) The use of the object of Claim 1 in an electrochemical cell for the generation of energy, such as a fuel cell.
- 5.) The use of the object of Claim 2 in an electrochemical cell for the generation of energy, such as a fuel cell.
- 6.) The use of the object of Claim 3 in an electrochemical cell for the generation of energy, such as a fuel cell.
- 7.) The object of Claim 1 where the assembly taught therein is permanently contained within a solid frame, such frame serving to seal the fluid within the structure from the outside and direct the flows of the fluids inside the structure.
- 8.) The object of Claim 7 with the frame fabricated from metal.
- 9.) The object of Claim 1 with the fluid barrier extending sufficiently far beyond the fluid distribution area to serve as a seating area for polymeric gaskets which are used to seal the structure.
- 10.) A fluid separator plate useful in electrochemical cells having a polymeric barrier formed within a metal component formed to insure the even distribution of a fluid flow over the surface of the separator.
- 11.) The use of the object of Claim 10 in an electrochemical cell for the generation of energy, such as a fuel cell.
- 12.) The object of Claim 10 where the assembly taught therein is permanently contained within a solid frame, such frame serving to seal the fluid within the structure from the outside and direct the flows of the fluids inside the structure.
- 13.) The object of Claim 10 where the formed metal component extends sufficiently far beyond the fluid distribution region that a sealing barrier can be constructed by filling the metal completely with polymer.
- 14.) The object of Claim 13 where the polymer extends beyond the metal.

- 15.) The object of Claim 10 where the polymer used to fabricate the barrier is selected to impart additional properties to the barrier.
- 16.) The object of Claim 15 where the property is the ability to permit the transport of water while preventing the transport of gas.
- 17.) The object of Claim 10 where the structure includes provision for the transport of a fluid within the plane of the separator plate.
- 18.) The object of Claim 17 where the fluid transported within the plane serves to remove heat from the separator.
- 19.) The object of Claim 10 where the metal component is a continuous structure, highly porous in all directions.
- 20.) The object of Claim 19 where the metal structure is that commonly known as foamed metal.
- 21.) The object of Claim 20 where a set of channels have been inscribed into the surface of the metal to improve the fluid distribution.
- 22.) The object of Claim 21 where the channels are inscribed to less than half of the total distance from the surface to the fluid barrier.
- 23.) The object of Claim 22 where the channels are arranged in two sets, one set beginning at one edge of the piece and extending at least half the distance to the opposite edge, and the second set beginning from the opposite edge and extending at least half the distance to the first edge, with the channels arranged such that they do not intersect, and preferably in an alternating, or interdigitated, pattern.

References

1. D. S. Watkins, *Fuel Cell Systems*, Blomen and Mugerwa, eds., Plenum, New York, (1993), 493-530.
2. R. K. A. M. Mallant, F. G. H. Koene, C. W. G. Verhoeve and A. Ruiter, *Prog. and Abstr., 1994 Fuel Cell Seminar*, (1994), 503-506.

Signatures

We, the undersigned, acknowledge the preceding 24 pages to be a true account of our invention:

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Eric T. Clarke
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